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MICROPHYSICAL STUDIES OF NOCTILUCENT CLOUDS

Final Technical Report

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MICROPHYSICAL STUDIES OF NOCTILUCENT CLOUDS

I. BACKGROUND

Very little is understood about the microphysics of noctilucent clouds. It is not known whether the ice crystals comprising them are of cubic habit, as has been suggested, or whether they are hexagonal, as in tropospheric clouds. It has not been established whether the crystal habit and the crystal growth rates are influenced or controlled by the nuclei present in the upper atmosphere, or whether temperature and humidity conditions play the dominant role in that respect as in the troposphere. It is clear, however, that the size and shape of the individual crystals determine their optical properties and therefore determine how noctilucent clouds interact with transmissions of electromagnetic waves.

Our experiments have been aimed at revealing the nature of the microphysical processes that lead to the formation of noctilucent clouds, and at revealing the nature of the particles that comprise them.

II. EXPERIMENTS AND RESULTS

The primary objectives of the investigation were to determine the habit of ice crystals formed at low atmospheric temperatures and pressures, and to investigate their nucleation kinetics, i.e., whether the nucleation process is stochastic as when ice is nucleated in the liquid phase, or whether it is deterministic. An additional objective was to investigate what influence, if any, the presence of foreign substances, vapors and particulate matter, might exert upon nucleation kinetics, growth rates and crystal habit. For this purpose it was attempted to modify and adapt well known techniques devised for studies of ice crystals under tropospheric conditions for use under noctilucent cloud conditions.

Our initial scheme was the adaptation (shown schematically in Figure 1) of the conventional thermal diffusion chamber. The chamber in this scheme consists of an evacuated glass capillary of 0.1 mm inner diameter, that has been loaded with a bead of water and sealed at both ends. The capillary is partially inside insulated copper tubes that have been wound with heater coils such that the temperature along the capillary can be controlled. Part of the capillary extends from the tubes and is illuminated in order to facilitate microscope observations of events inside. A heater coil is mounted beneath this extending part so its temperature can be controlled as well. With the water bead serving as a reservoir in the hidden end of the capillary, the saturation ratio in the extending end can be controlled by manipulating the temperature along the length of the capillary. supersaturation is attained in the viewing section by cooling it relative to the temperature at the reservoir; undersaturation is obtained by heating it. With the system immersed in a thermostated, low-temperature environment chamber, crystals can be nucleated at will in the viewing section of the capillary, grown to any desirable size, and destroyed by simple manipulation of the temperature difference between the capillary's ends.

The initial runs of the evacuated capillary system at tropospheric temperatures were extremely encouraging. With the aid of a microscope we could watch condensation of liquid water onto the capillary walls and its subsequent sudden freezing. We could observe a generally decreasing delay time between condensation of liquid and freezing as the temperature was lowered, but at temperatures between -20°C and -30°C it became impossible to discern this delay. Below this range of temperatures, isolated, single crystals could be nucleated and grown to recognizable shapes, provided supersaturation was maintained low. Too high supersaturation resulted in nucleation of ice all over the capillary walls and the formation of a continuous sheet of ice. Commonly, isolated hexagonal bullets or plates could be observed.

However, when the temperature was lowered towards noctilucent cloud temperature (-150°C), the growth rates of the crystals became extremely low. It became impossible in a days time to grow and nurture isolated crystals to optically observable sizes. Only by means of large supersaturations could any ice at all be observed, but then the crystals had nucleated in great numbers and grown into each other so that no crystal structure could be perceived. This applied to temperatures as high as about -70°C.

At temperatures between -60°C and -70°C crystal habit could be perceived after a few hours of crystal growth. In this temperature range crystals exhibiting cubic or rectangular habits were frequently observed. Images of some of these were recorded on video. Figure 2 shows traces of such video images obtained at -30°C and -62°C. The former is cubic in shape; the latter hexagonal.

As a result of the very much reduced growth rates of ice crystals at noctilucent cloud temperatures, it became clear that utilization of the technique of repeated nucleation under constant conditions to determine nucleation kinetics was unfeasible. At the low supersaturations required to limit the number of nuclei or nucleation sites in each event to one or only a few growth rates were so low that a considerable time was needed to allow the crystals formed to grow to detectable sizes. Even at tropospheric temperatures we found this growth period to constitute most of the observed time intervals between nucleation events. At noctilucent cloud temperatures the saturation vapor pressure may be as much as 10 orders of magnitude lower than at tropospheric temperatures, while the diffusion rate may be increased by 5 orders of magnitude due to the lower pressure. For a given supersaturation, ice crystal growth rates may thus be as much as 5 orders of magnitude lower in noctilucent clouds than in tropospheric clouds. A crystal that might be detected a few seconds after its nucleation at tropospheric temperatures may, therefore, have to be nurtured invisible and undetected by any optical means for up to a few days under noctilucent cloud conditions while temperature and humidity are maintained constant. A meaningful dataset would have to be based on hundreds, or preferably, thousands of such events, and thus would require a continuous run for a duration of months or even years. Clearly, under these conditions, the adaptation of conventional nucleation techniques to noctilucent cloud conditions was impracticable. Our observations indicated that at temperatures below -70°C growth rates became prohibitively low for any studies involving repetitive formation and growth of ice crystals.

We decided, nevertheless, to attempt to obtain data on repetitious nucleation of ice directly from the vapor phase, if not at noctilucent cloud temperatures, then at as low temperatures as possible. But several technical problems arose in our attempts to electronically detect ice crystals as they formed inside the capillary for timing of events and automatic control of the saturation ratio. The main problem appeared to be lack of contrast between the ice crystals and the glass walls on which they formed, but also the fact that repeated nucleation events did not necessarily always take place at the same site. Therefore, we devised another scheme in which the crystals were no longer encapsulated in glass.

The second scheme is illustrated in Figure 3. Supersaturation is attained by mixing two air masses of different temperatures but both at water vapor saturation. The mixing is accomplished in a jet emanating from a humidity chamber through a small nozzle into a larger constant temperature chamber in which the air is also saturated. The supersaturation is controlled by varying the temperature of the humidity chamber.

The target on which ice crystals were grown was a 10 um diameter tantalum wire connected to a current source through a relay switch. Crystal formation on the wire was detected by a photodiode connected to a comparator circuit that closed the relay when the scattered light from the crystal had reached a certain intensity, thus causing a current to flow through the wire, heating the crystal and causing it to evaporate. At the same time a time signal was generated and the nucleation cycle started anew.

Figure 4 shows results obtained at a temperature of about $-45^{\circ}C$ and temperature difference between the two air masses of about $2^{\circ}C$. We see a large scatter in the elapsed time intervals between events. In runs in which the temperature difference was larger, this scatter disappeared. However, upon closer analysis of the data, it becomes evident that it does not conform to a Poissons distribution as would be expected of a stochastic process, but appears distributed more in a Gaussian sense about a certain mean. This prompted a closer look at the signals from the photodiode. When recorded directly onto a strip chart recorder it became evident that all the scatter in Figure 4 was due to variations in crystal growth rates, and not due to nucleation delay. A trace of such a strip charge record is shown in Figure 5, along with a trace of difference in temperature between the two air masses.

III. DISCUSSION

We conclude from our experiments that the ice particles present in noctilucent clouds may often exhibit cubic habit. It appears even possible that high tropospheric ice crystals may occasionally exhibit such habit. That would be consistent with rare, but reliable observations of the Scheiner's halo.

In the case of noctilucent clouds, however, it is doubtful that the crystal habit is after all of much practical importance, because due to low growth rates and limited duration of supersaturation at the mesopause, the crystals must be very small. Ice crystals of dimensions much smaller than the wavelength of

transmitted electromagnetic waves interact with the waves as Rayleigh scatterers, which allows for fairly simple theoretical treatments of the interactions. No refraction or surface reflection would be occurring. It appears that even after a few days of growth noctilucent cloud particles are still only a fraction of a micron in diameter. That is less than or equivalent to the wavelength of visible light.

Although our experiments failed to demonstrate that ice nucleation from the gas phase is a stochastic process, the contrary (i.e., that the process is deterministic) does not necessarily follow. It is possible that even at -45°C vapor was first condensing as liquid on our target which then froze within an unperceivable time interval. It is also possible that our target, even though it was only 10 um in diameter, contained many active nucleation sites along the length (5 mm) exposed to the supersaturated air mixture. If this were the case, collectively these sites could have had a large probability of nucleation associated with them, although each individual site might have had a certain, smaller probability associated with it. Actually, our observation of variable crystal growth rates under the same conditions could be explained by progressive nucleation at some of these sites as growth progressed at others.

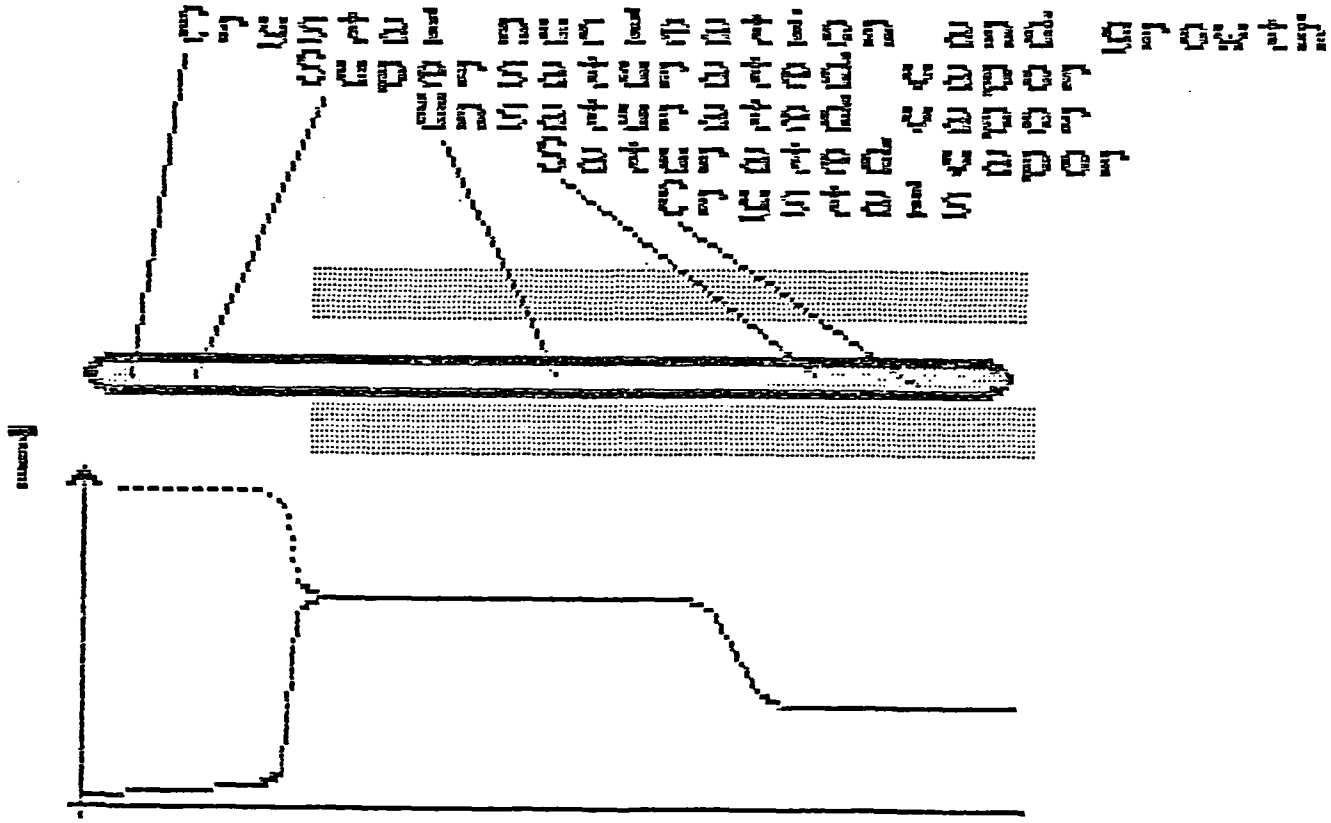
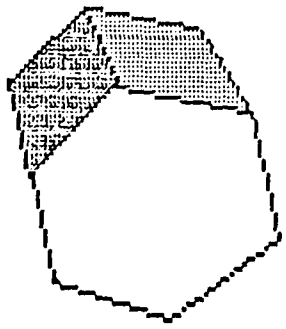


FIGURE 1

Crystal Habits Observed in Low Pressure Chambers

1.

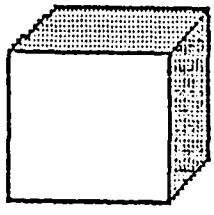


T = - 30C

Only hexagonal shapes are seen

T = -62C

2.



Cubic shapes are frequently seen

FIGURE 2

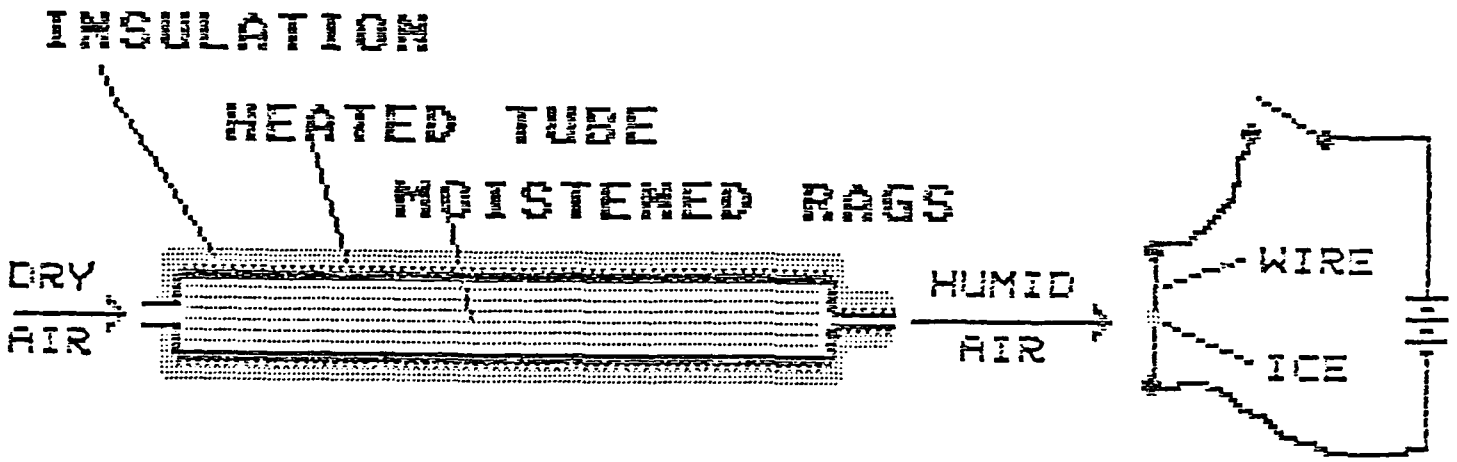

INSULATION
HEATED TUBE
MOISTENED RAGS
DRY AIR
HUMID AIR
WIRE
ICE

FIGURE 3

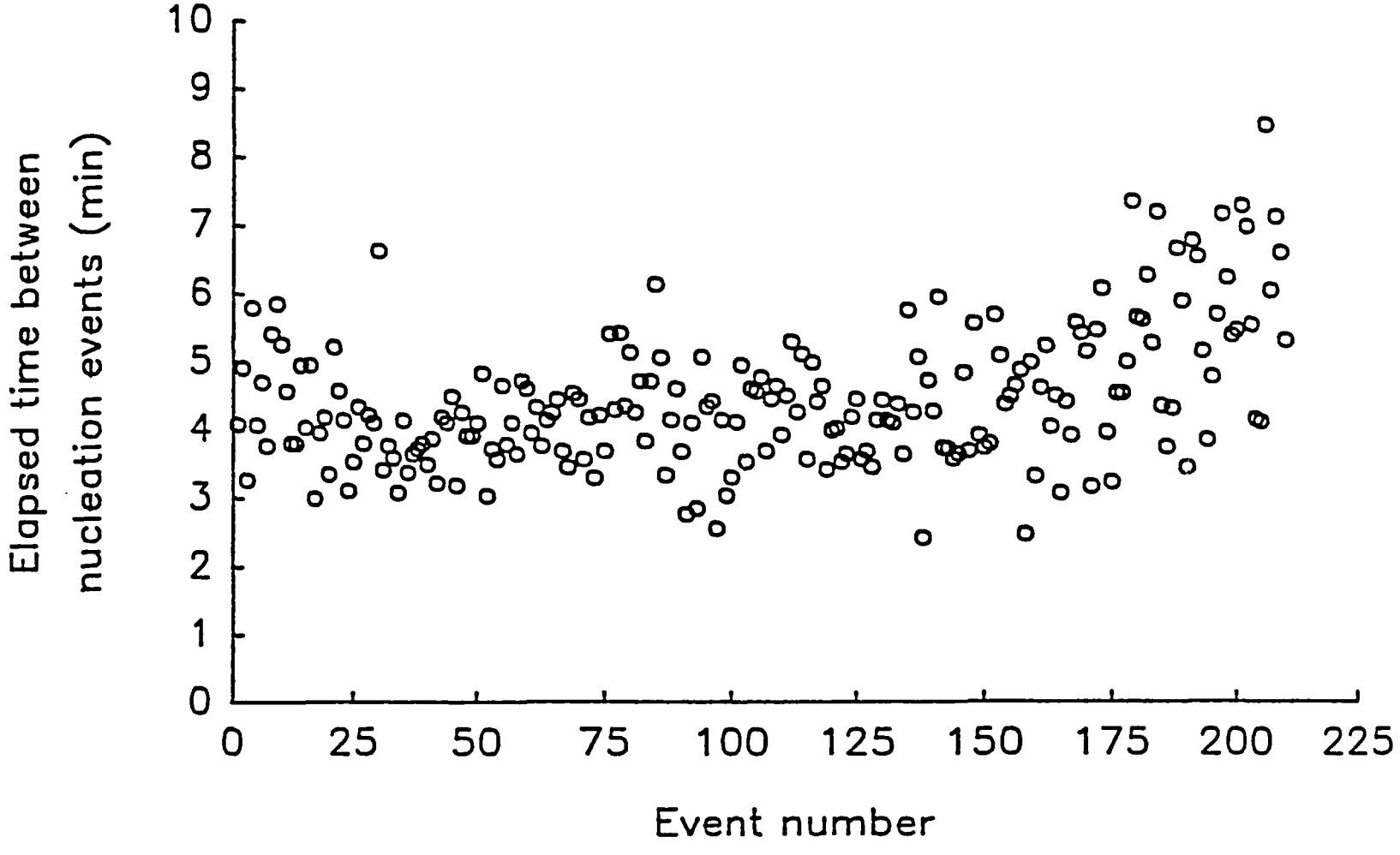

Elapsed time between nucleation events (min)
10
9
8
7
6
5
4
3
2
1
0
0
25
50
75
100
125
150
175
200
225
Event number

FIGURE 4

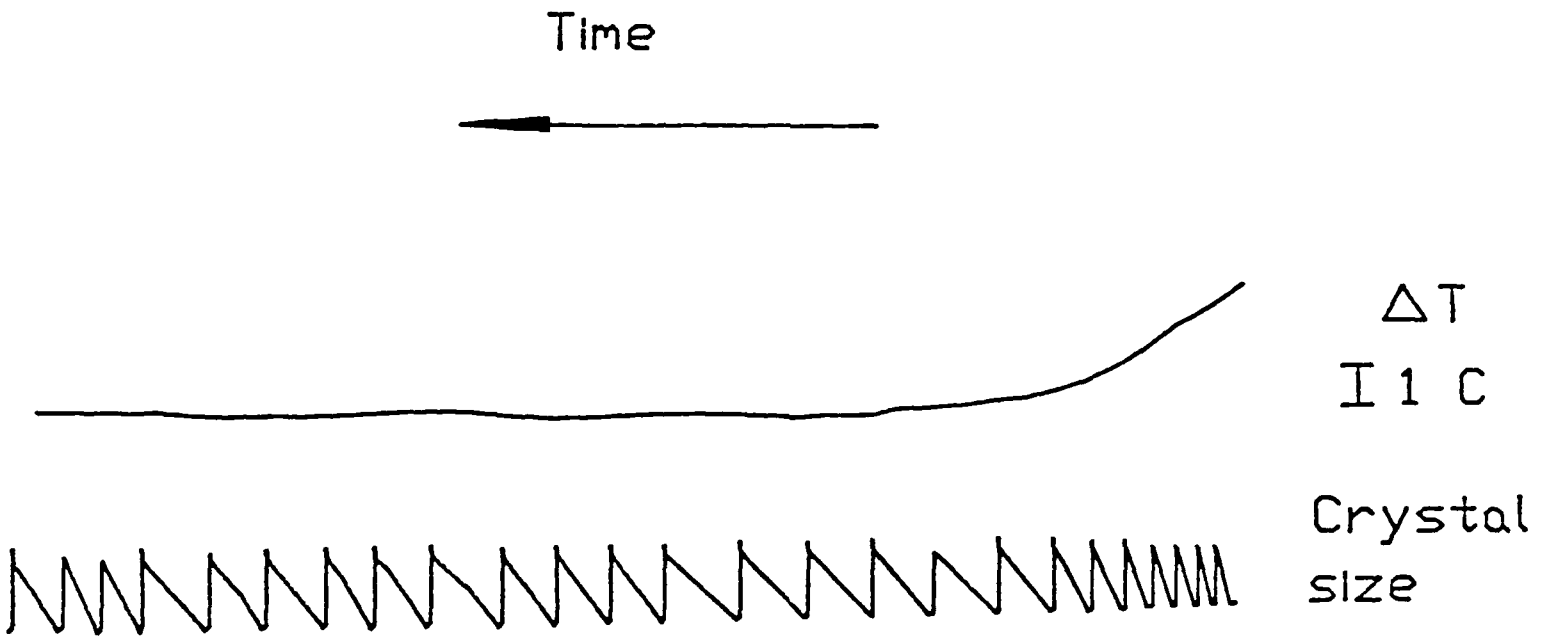


FIGURE 5